Ioffe Institute
6th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 22–26, 1998.

Control of the quantum dot energy by a photon: Observation of two-exciton and three-exciton states in quantum dots

Yasuaki Masumoto†‡ and Michio Ikezawa†
† Institute of Physics, University of Tsukuba, Tsukuba 305-8571, Japan
‡ Single Quantum Dot Project, ERATO, JST, Tsukuba Research Consortium, Tsukuba 300-2635, Japan

Abstract. Antibonding two-exciton and three-exciton states were observed in CuCl quantum dots. This observation demonstrates the controllability of the quantum dot energy by a photon.

A semiconductor quantum dot is so small that its energy is changed as a result of the absorption of a photon. Let us consider three cases. The first case is the case where any excitons are not in a quantum dot. The second case is the case where an exciton is in it. The third case is the case where a biexciton is in it. When a photon creates an exciton or an additional exciton in a dot, an exciton, two excitons and three excitons will be in the dots depending on three cases. In a bulk crystal, two excitons and three excitons can be located apart without any interaction. Then the photon energies to create an exciton and additional exciton(s) are the same. However, in a quantum dot, excitons are forced to interact with each other because of the short distance between them. If the photon energy which creates an exciton or an additional exciton in a dot depends on three cases considerably, the energy of the quantum dot is controllable by a photon.

One of two-exciton states is known as the biexciton ground state in bulk crystals. It is also observed in quantum dots and the size dependence is studied [1]. The biexciton ground state is considered as a bonding state of two-exciton states. However, an additional two-exciton state, in other words, the biexciton excited state in quantum dots has been predicted by the theory [2]. This state, an antibonding state of two excitons, is not present in the bulk crystal, while it is expected to be present uniquely in quantum dots. It is expected to depend on the size of the dots strongly. The purpose of this paper is to demonstrate the antibonding two-exciton and three-exciton states in the quantum dots [3, 4].

For the observation of two-exciton and three-exciton states, we used a model quantum dot in the weak confinement regime, a CuCl quantum dot embedded in a NaCl crystal. The picosecond-pump-and-femtosecond-probe spectroscopy was used for the investigation. The laser system we used was a 1 kHz, 300 fs Ti:sapphire regenerative amplifier system. The second harmonics of the output was spectrally narrowed passing through a spectral filter stage made of a grating and was used as the pump pulses. The spectral width was 1.7 meV which was suitable for the size-selective excitation of quantum dots, but the temporal width was elongated to 1.2 ps. The probe light was a white continuum in the femtosecond range. The time-resolved absorption spectra were measured by means of a 25 cm monochromator and a liquid nitrogen cooled charge-coupled device.

The Z_3 exciton and $Z_{1,2}$ exciton absorption structures observed in the upper part of Fig. 1 are shifted to the higher energy side compared to those of the bulk crystal due

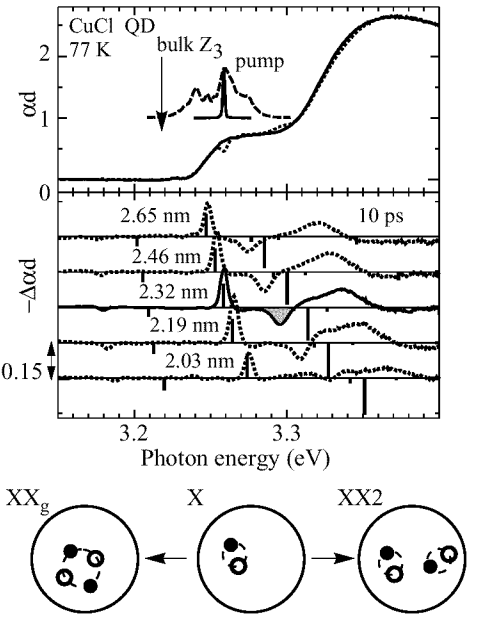

Fig 1. Upper figure: The solid line shows absorption spectrum of CuCl quantum dots embedded in a NaCl crystal at 77 K, while a dashed line represents that at 10 ps after photoexcitation. Lower figure: The solid line shows differential absorption spectrum corresponding to the upper figure, while dashed lines are those for different excitation photon energies. The radii of quantum dots which were excited are 2.65 nm, 2.46 nm, 2.32 nm, 2.19 nm and 2.03 nm from top to bottom, respectively. Thick solid bars are theoretical results normalized at the spectral hole. In the inset, filtered and unfiltered pump spectra are shown by a solid line and a dashed line, respectively. Two excitation processes of the two-exciton states are illustrated.

to the quantum confinement effect. The Z_3 exciton energy of bulk CuCl is shown by a downward arrow. According to the well-known relation between the quantum confined exciton energy and the quantum dot radius [5], the pump pulse energy size-selectively excite the quantum dots of the corresponding radius. The differential absorption spectrum in the lower part of Fig. 1 consists of a spectral hole at the pump photon energy and two induced absorption structures at both sides (3.180 eV, 3.296 eV) of the spectral hole.

Differential absorption spectra for four different pump photon energies are also shown at the lower part of Fig. 1. Two induced absorption bands shift with the change of the excitation photon energy. The theoretical induced absorption spectrum from the exciton ground state is shown by thick solid bars in Fig. 1 [4]. The energy shifts of strong bands show good correspondence with experiments. Furthermore, the relative strengths of the induced absorption lines and the spectral hole are reproduced quite well by the theory. The strong peak above the pump energy can be assigned to the induced absorption transition to antibonding two-exciton ($XX2$) state [2].

The energies of the spectral hole and the induced absorption bands are plotted in Fig. 2 as a function of the excitation photon energy. The solid circles show spectral

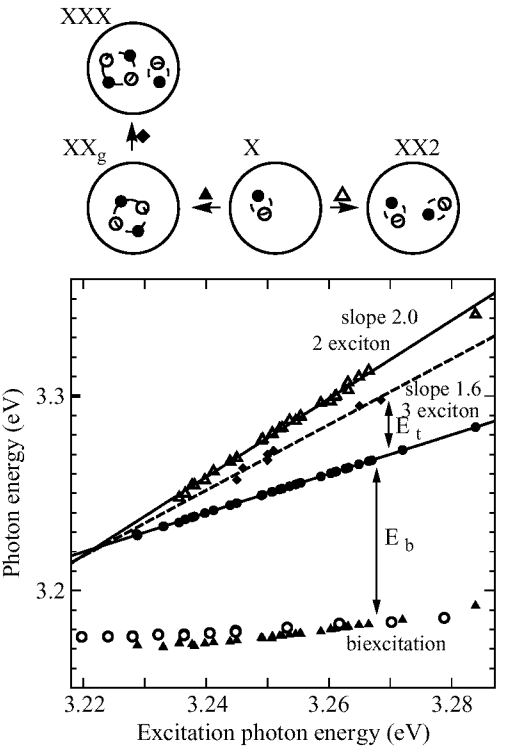

Fig 2. Excitation energy dependence of the structures appearing in the differential absorption spectrum. The solid circles represent spectral hole energies, and they are on a line of slope 1.0. Open (solid) triangles show the energy of the induced absorption located at the higher (lower) energy side of the spectral hole. Open circles indicate the energy of the induced absorption measured by nanosecond pump-and-probe method in Ref. [1]. Solid diamonds correspond to additional induced absorption under high-density excitation. E_b denotes the biexciton binding energy defined by $2E_X - E_{XX}$ and E_t shows $E_{XXX} - E_{XX} - E_X$. Then the binding energy of three-exciton state defined by $3E_X - E_{XXX}$ is given by $E_b - E_t$. The excitation processes of two-exciton states and three-exciton state are illustrated at the top part.

hole energies, and they are on a line of slope 1.0, since their energy coincides with the excitation photon energy. Open (solid) triangles exhibit the energies of the induced absorption located at the higher (lower) energy side. Open circles show the energies of induced absorption measured previously by nanosecond pump-and-probe method [1] which was identified as the transition from the exciton (X) to the biexciton ground state (XXg).

The solid line through the open triangles has a slope of 2.0. Antibonding of two excitons naturally explains the slope [4]. This line crosses the line of slope 1.0 near the Z_3 exciton energy of bulk CuCl. Furthermore, the spectral hole and the induced absorption on the higher energy side exhibit almost the same temporal evolution with a decay time constant of 480 ps. This fact is reasonable, because the induced absorption on the higher energy side arises from the transition from excitons pre-excited in the

quantum dots.

The three-exciton state (XXX) was found as an additional induced absorption band by two independent methods employing high-density excitation and two-color excitation. Then the biexciton luminescence became observable. In the two-color pump-and-probe method, the energy of the second pump pulse was tuned to the induced absorption caused by the first pump pulse. This combination produces two-exciton state effectively only in quantum dots of particular size and enables us to observe the induced absorption to three-exciton states. Excitation energy dependence of this induced absorption is shown in Fig. 2 by solid diamonds. The fitted line has a slope of 1.7 and this line also crosses the other two lines near the bulk Z_3 exciton energy. An anti-bonding combination of a biexciton and an exciton explains the slope of 1.7 [4]. The decay time of the additional induced absorption corresponds to the biexciton luminescence lifetime of 70 ps. This shows that the additional induced absorption can be assigned to transition from the biexciton ground state to a three-exciton state.

In conclusion, we found two-exciton and thre-exciton states in quantum dots in the weak confinement regime by using time-resolved size-selective pump and probe technique. These experiments open a new direction of research of many-exciton states in confined systems and demonstrate the controllability of the quantum dot energy by a photon.

The authors acknowledge Dr. S. V. Nair and Dr. T. Takagahara for the theoretical support throughout this work.

References

[1] Y. Masumoto, S. Okamoto, and S. Katayanagi. *Phys. Rev.* B **50** 18658 (1994).
[2] S. V. Nair and T. Takagahara. *Phys. Rev.* B **53** R10516 (1996); *Phys. Rev.* B **55** 5153 (1997).
[3] M. Ikezawa and Y. Masumoto. *Jap. J. Appl. Phys.* **36** 4191 (1997).
[4] M. Ikezawa, Y. Masumoto, T. Takagahara and S.V. Nair. *Phys. Rev. Lett.* **79** 3522 (1997).
[5] T. Itoh, Y. Iwabuchi, and M. Kataoka. *Phys. Status Solidi* B **145** 567 (1988).